

VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS

PCT

INTERNATIONALER RECHERCHENBERICHT

(Artikel 18 sowie Regeln 43 und 44 PCT)

Aktenzeichen des Anmelders oder Anwalts 1998/F151 PCT	WEITERES VORGEHEN siehe Mitteilung über die Übermittlung des internationalen Recherchenberichts (Formblatt PCT/ISA/220) sowie, soweit zutreffend, nachstehender Punkt 5
Internationales Aktenzeichen PCT/EP 99/ 09831	Internationales Anmeldedatum (Tag/Monat/Jahr) 11/12/1999
	(Frühestes) Prioritätsdatum (Tag/Monat/Jahr) 25/12/1998
Anmelder AXIVA GMBH	

Dieser internationale Recherchenbericht wurde von der Internationalen Recherchenbehörde erstellt und wird dem Anmelder gemäß Artikel 18 übermittelt. Eine Kopie wird dem Internationalen Büro übermittelt.

Dieser internationale Recherchenbericht umfaßt insgesamt 3 Blätter.

☒ Darüber hinaus liegt ihm jeweils eine Kopie der in diesem Bericht genannten Unterlagen zum Stand der Technik bei.

1. Grundlage des Berichts

a. Hinsichtlich der **Sprache** ist die internationale Recherche auf der Grundlage der internationalen Anmeldung in der Sprache durchgeführt worden, in der sie eingereicht wurde, sofern unter diesem Punkt nichts anderes angegeben ist.

☐ Die internationale Recherche ist auf der Grundlage einer bei der Behörde eingereichten Übersetzung der internationalen Anmeldung (Regel 23.1 b)) durchgeführt worden.

b. Hinsichtlich der in der internationalen Anmeldung offenbarten **Nucleotid- und/oder Aminosäuresequenz** ist die internationale Recherche auf der Grundlage des Sequenzprotokolls durchgeführt worden, das

☐ in der internationalen Anmeldung in Schriftlicher Form enthalten ist.

☐ zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.

☐ bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.

☐ bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.

☐ Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.

☐ Die Erklärung, daß die in computerlesbarer Form erfaßten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

2. ☐ Bestimmte Ansprüche haben sich als nicht recherchierbar erwiesen (siehe Feld I).

3. ☐ Mangelnde Einheitlichkeit der Erfindung (siehe Feld II).

4. Hinsichtlich der Bezeichnung der Erfindung

☒ wird der vom Anmelder eingereichte Wortlaut genehmigt.

☐ wurde der Wortlaut von der Behörde wie folgt festgesetzt:

5. Hinsichtlich der Zusammenfassung

☒ wird der vom Anmelder eingereichte Wortlaut genehmigt.

☐ wurde der Wortlaut nach Regel 38.2b) in der in Feld III angegebenen Fassung von der Behörde festgesetzt. Der Anmelder kann der Behörde innerhalb eines Monats nach dem Datum der Absendung dieses internationalen Recherchenberichts eine Stellungnahme vorlegen.

6. Folgende Abbildung der Zeichnungen ist mit der Zusammenfassung zu veröffentlichen: Abb. Nr. 1

☒ wie vom Anmelder vorgeschlagen

☐ weil der Anmelder selbst keine Abbildung vorgeschlagen hat.

☐ weil diese Abbildung die Erfindung besser kennzeichnet.

☐ keine der Abb.

A. KLASSIFIZIERUNG DES ANMELDUNGSGEGENSTANDES

IPK 7 C08J5/22 C25B9/00 H01M8/10

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

B. RECHERCHIERTE GEBIETE

Recherchierter Mindestprüfstoff (Klassifikationssystem und Klassifikationssymbole)

IPK 7 C08J C25B H01M

Recherchierte aber nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)

C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie°	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
X	WO 98 14505 A (HOECHST CELANESE CORP ;SANSONE MICHAEL J (US); MARIKAR FARUQ (US);) 9. April 1998 (1998-04-09) Beispiel 1 Seite 7, Zeile 12 -Seite 8, Zeile 2 Ansprüche 1,10,12 ---	1-7
A	WAINRIGHT J S ET AL: "ACID-DOPED POLYBENZIMIDAZOLES: A NEW POLYMER ELECTROLYTE" JOURNAL OF THE ELECTROCHEMICAL SOCIETY,US,ELECTROCHEMICAL SOCIETY. MANCHESTER, NEW HAMPSHIRE, Bd. 142, Nr. 7, 1. Juli 1995 (1995-07-01), Seiten 121-123, XP000770321 ISSN: 0013-4651 in der Anmeldung erwähnt das ganze Dokument --- -/--	1

☒ Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen☒ Siehe Anhang Patentfamilie

° Besondere Kategorien von angegebenen Veröffentlichungen :

- "A" Veröffentlichung, die den allgemeinen Stand der Technik definiert, aber nicht als besonders bedeutsam anzusehen ist
- "E" älteres Dokument, das jedoch erst am oder nach dem internationalen Anmeldedatum veröffentlicht worden ist
- "L" Veröffentlichung, die geeignet ist, einen Prioritätsanspruch zweifelhaft erscheinen zu lassen, oder durch die das Veröffentlichungsdatum einer anderen im Recherchenbericht genannten Veröffentlichung belegt werden soll oder die aus einem anderen besonderen Grund angegeben ist (wie ausgeführt)
- "O" Veröffentlichung, die sich auf eine mündliche Offenbarung, eine Benutzung, eine Ausstellung oder andere Maßnahmen bezieht
- "P" Veröffentlichung, die vor dem internationalen Anmeldedatum, aber nach dem beanspruchten Prioritätsdatum veröffentlicht worden ist

"T" Spätere Veröffentlichung, die nach dem internationalen Anmeldedatum oder dem Prioritätsdatum veröffentlicht worden ist und mit der Anmeldung nicht kollidiert, sondern nur zum Verständnis des der Erfindung zugrundeliegenden Prinzips oder der ihr zugrundeliegenden Theorie angegeben ist

"X" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann allein aufgrund dieser Veröffentlichung nicht als neu oder auf erfinderischer Tätigkeit beruhend betrachtet werden

"Y" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als auf erfinderischer Tätigkeit beruhend betrachtet werden, wenn die Veröffentlichung mit einer oder mehreren anderen Veröffentlichungen dieser Kategorie in Verbindung gebracht wird und diese Verbindung für einen Fachmann naheliegend ist

"&" Veröffentlichung, die Mitglied derselben Patentfamilie ist

Datum des Abschlusses der internationalen Recherche

11. Mai 2000

Absendedatum des internationalen Recherchenberichts

22/05/2000

Name und Postanschrift der Internationalen Recherchenbehörde
Europäisches Patentamt, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Bevollmächtigter Bediensteter

Niaounakis, M

C.(Fortsetzung) ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
A	<p>WANG J -T ET AL: "A H2/O2 FUEL CELL USING ACID DOPED POLYBENZIMIDAZOLE AS POLYMER ELECTROLYTE"</p> <p>ELECTROCHIMICA ACTA,GB,ELSEVIER SCIENCE PUBLISHERS, BARKING, Bd. 41, Nr. 2, 1. Februar 1996 (1996-02-01), Seiten 193-197, XP000770320 ISSN: 0013-4686 Seite 193, linke Spalte, Zeile 1 -Seite 195, linke Spalte, Zeile 8</p> <p>---</p>	1
A	<p>US 4 795 536 A (YOUNG PING ET AL) 3. Januar 1989 (1989-01-03) Spalte 7, Zeile 35 -Spalte 8, Zeile 40 Anspruch 1</p> <p>---</p>	1
A	<p>WO 97 37396 A (UNIV CASE WESTERN RESERVE) 9. Oktober 1997 (1997-10-09) in der Anmeldung erwähnt Ansprüche 1-4</p> <p>---</p>	1
A	<p>WO 96 13872 A (UNIV CASE WESTERN RESERVE) 9. Mai 1996 (1996-05-09) in der Anmeldung erwähnt Ansprüche 34-36</p> <p>---</p>	1
A	<p>US 5 599 639 A (OGATA NAOYA ET AL) 4. Februar 1997 (1997-02-04) Ansprüche 1,2,9</p> <p>-----</p>	1

INTERNATIONAL SEARCH REPORT

Int'l Application No

PCT/EP 99/09831

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08J5/22 C25B9/00 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J C25B H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 14505 A (HOECHST CELANESE CORP ;SANSONE MICHAEL J (US); MARIKAR FARUQ (US);) 9 April 1998 (1998-04-09) example 1 page 7, line 12 -page 8, line 2 claims 1,10,12	1-7
A	WAINRIGHT J S ET AL: "ACID-DOPED POLYBENZIMIDAZOLES: A NEW POLYMER ELECTROLYTE" JOURNAL OF THE ELECTROCHEMICAL SOCIETY,US,ELECTROCHEMICAL SOCIETY. MANCHESTER, NEW HAMPSHIRE, vol. 142, no. 7, 1 July 1995 (1995-07-01), pages 121-123, XP000770321 ISSN: 0013-4651 cited in the application the whole document	1

-/-

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"Z" document member of the same patent family

Date of the actual completion of the international search

11 May 2000

Date of mailing of the international search report

22/05/2000

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
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Authorized officer

Niaounakis, M

INTERNATIONAL SEARCH REPORT

Int. Application No.

PCT/EP 99/09831

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WANG J -T ET AL: "A H ₂ /O ₂ FUEL CELL USING ACID DOPED POLYBENZIMIDAZOLE AS POLYMER ELECTROLYTE" ELECTROCHIMICA ACTA, GB, ELSEVIER SCIENCE PUBLISHERS, BARKING, vol. 41, no. 2, 1 February 1996 (1996-02-01), pages 193-197, XP000770320 ISSN: 0013-4686 page 193, left-hand column, line 1 -page 195, left-hand column, line 8	1
A	US 4 795 536 A (YOUNG PING ET AL) 3 January 1989 (1989-01-03) column 7, line 35 -column 8, line 40 claim 1	1
A	WO 97 37396 A (UNIV CASE WESTERN RESERVE) 9 October 1997 (1997-10-09) cited in the application claims 1-4	1
A	WO 96 13872 A (UNIV CASE WESTERN RESERVE) 9 May 1996 (1996-05-09) cited in the application claims 34-36	1
A	US 5 599 639 A (OGATA NAOYA ET AL) 4 February 1997 (1997-02-04) claims 1,2,9	1

INTERNATIONAL SEARCH REPORT

information on patent family members

Int'l. Application No

PCT/EP 99/09831

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 9814505	A	09-04-1998	AU 4893997 A EP 0954544 A	24-04-1998 10-11-1999
US 4795536	A	03-01-1989	NONE	
WO 9737396	A	09-10-1997	US 5716727 A AU 2603997 A EP 0907977 A US 6025085 A	10-02-1998 22-10-1997 14-04-1999 15-02-2000
WO 9613872	A	09-05-1996	US 5525436 A AU 4017595 A EP 0787369 A JP 11503262 T	11-06-1996 23-05-1996 06-08-1997 23-03-1999
US 5599639	A	04-02-1997	JP 9073908 A	18-03-1997

PATENT COOPERATION TREATY

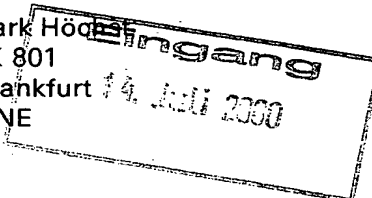
PCT

NOTICE INFORMING THE APPLICANT OF THE
COMMUNICATION OF THE INTERNATIONAL
APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

From the INTERNATIONAL BUREAU

To:

AXIVA GMBH
Patente
Industriepark Höchst
Gebäude K 801
D-65926 Frankfurt
ALLEMAGNE

e Techn. Sachb. ord. OK

Date of mailing (day/month/year) 06 July 2000 (06.07.00)		IMPORTANT NOTICE	
Applicant's or agent's file reference 1998/F151 PCT			
International application No. PCT/EP99/09831	International filing date (day/month/year) 11 December 1999 (11.12.99)	Priority date (day/month/year) 25 December 1998 (25.12.98)	
Applicant AXIVA GMBH et al		MIS/Datenpflege Erledigung:	

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
CN,JP,KR,US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
BR,CA,CZ,EP,MX,PL,RU,SG

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
06 July 2000 (06.07.00) under No. WO 00/39202

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a **demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

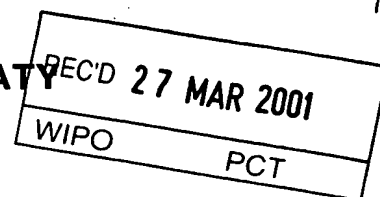
Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38



INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 1998/F151	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP99/09831	International filing date (day/month/year) 11/12/1999	Priority date (day/month/year) 25/12/1998
International Patent Classification (IPC) or national classification and IPC C08J5/22		
Applicant (AXIVA GMBH et al.) CELANESE VENTURES GMBH		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.


2. This REPORT consists of a total of 5 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 06/07/2000	Date of completion of this report 23.03.2001
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Puttins, U Telephone No. +49 89 2399 8661



INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP99/09831

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).):*

Description, pages:

1-13 as originally filed

Claims, No.:

1-7 as originally filed

Drawings, sheets:

1/3-3/3 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/09831

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims 1-7
	No: Claims
Inventive step (IS)	Yes: Claims
	No: Claims 1-7
Industrial applicability (IA)	Yes: Claims 1-7
	No: Claims

- 2. Citations and explanations**
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

R Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty and inventive step; citations and explanations supporting such statement

Reference is made to the following document:

D1: WO 98 14505 A (HOECHST CELANESE CORP ;SANSONE MICHAEL J (US); MARIKAR FARUQ (US);) 9. April 1998 (1998-04-09)

1. Novelty (Art.33(2) PCT):

The subject-matter of present claims 1 to 7 is considered novel over the prior art documents cited in the International Search Report and the application, since none of the documents discloses a process for the manufacture of a polyelectrolyte membrane, comprising the step of immersing a basic polymer in a strong acid at a concentration of six or more strong acid molecules per polymer repeating unit at a temperature of at least 30°C for a period of 5h or less.

2. Inventive Step (Art.33(3) PCT):

The subject-matter of present claims 1 to 7 is not based upon an inventive step for the following reasons:

Document D1, which is considered to represent the closest prior art, discloses a method for producing a polyelectrolyte membrane by immersing a basic polymer in a strong acid (cf.D1, claims 1, 10 and 12).

The method claimed in present claims differs from that disclosed in D1 only in that the immersing temperature is held of not less than 30°C, instead of 23°C as disclosed in the examples of D1.

Whilst this feature is sufficient to establish novelty with respect to document D1, it is, however, insufficient to establish the presence of an inventive step for the following reasons:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP99/09831

Compared with the disclosure of D1, the problem to be solved by the present application may be regarded as to provide a method for producing a polyelectrolyte membrane, whereby the immersion time should be reduced and the concentration of the protons on the membrane should be enhanced.

A skilled man who intends to solve this problem was aware from document D1 that the use of elevated temperatures in the immersion step would lead to an increase of the imbibition of the acid and the immersion time could be reduced by increasing the temperature (cf. D1, page 7, lines 12 to 26).

The skilled man would therefore regard it as a normal measure to carry out the immersion at an elevated temperature such as 30°C or higher in order to solve the problem posed.

This way of solving the above mentioned problem would be obvious to the skilled man. Therefore, claims 1 to 7 of the present application cannot be regarded as involving an inventive step.

Re Item VII

Certain defects in the international application

- 1) The "incorporation by reference" of the entire contents of other documents could lead to doubt as to the extent of the claims (cf. pages 2 and 5).
- 2) Contrary to the requirements of Rule 5.1(a)(ii) PCT, the relevant background art disclosed in document D1 is not mentioned in the description, nor is this document identified therein.

RECEIVING OFFICE REQUEST FORM
1998/F-151

PC

(4) pages
ANTRAG

Der Unterzeichnete beantragt, daß die vorliegende internationale Anmeldung nach dem Vertrag über die internationale Zusammenarbeit auf dem Gebiet des Patentwesens behandelt wird.

Anmeldeamt auszufüllen

PCT/EP 99 / 098 3 1
Internationales Aktenzeichen

(11. 12. 1999) 11 DEC 1999
Internationales Anmeldedatum

OFFICE EUROPEEN DES BREVETS
DEMANDE INTERNATIONALE PCT
Name des Anmeldeamts und "PCT International Application"

Aktenzeichen des Anmelders oder Anwalts (falls gewünscht)
(max. 12 Zeichen) 1998/F151 PCT

Feld Nr. I BEZEICHNUNG DER ERFINDUNG

Method for Producing Polymer Electrolyte Membrane and Fuel Cell

Feld Nr. II ANMELDER

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.)

Aventis Research & Technologies GmbH & Co KG

D-65926 Frankfurt am Main
Deutschland

☐ Diese Person ist gleichzeitig Erfinder

Telefonnr.: 069-305-4302

Telefaxnr.: 069-305-16350

Fernschreibnr.:

Staatsangehörigkeit (Staat): DE

Sitz oder Wohnsitz (Staat): DE

Diese Person ist Anmelder für folgende Staaten: ☐ alle Bestimmungsstaaten ☒ alle Bestimmungsstaaten mit Ausnahme der Vereinigten Staaten von Amerika ☐ nur die Vereinigten Staaten von Amerika ☐ die im Zusatzfeld angegebenen Staaten

Feld Nr. III WEITERE ANMELDER UND/ODER (WEITERE) ERFINDER

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.)

YAMAMOTO, Tetsu
9-401, Torimachi 24-chome
Kawagoe, Saitama 350-0044
Japan

Diese Person ist:

☐ nur Anmelder

☒ Anmelder und Erfinder

☐ nur Erfinder (Wird dieses Kästchen angekreuzt, so sind die nachstehenden Angaben nicht nötig.)

Staatsangehörigkeit (Staat): JP

Sitz oder Wohnsitz (Staat): JP

Diese Person ist Anmelder für folgende Staaten: ☐ alle Bestimmungsstaaten ☐ alle Bestimmungsstaaten mit Ausnahme der Vereinigten Staaten von Amerika ☒ nur die Vereinigten Staaten von Amerika ☐ die im Zusatzfeld angegebenen Staaten

☐ Weitere Anmelder und/oder (weitere) Erfinder sind auf einem Fortsetzungsblatt angegeben.

Feld Nr. IV ANWALT ODER GEMEINSAMER VERTRETER; ODER ZUSTELLANSCHRIFT

Die folgende Person wird hiermit bestellt/ist bestellt worden, um für den (die) Anmelder vor den zuständigen internationalen Behörden in folgender Eigenschaft zu handeln als: ☐ Anwalt ☐ gemeinsamer Vertreter.

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben.)

Aventis Research & Technologies GmbH & Co KG
Patent- und Lizenzabteilung,
Industriepark Höchst, Gebäude K 801
D-65926 Frankfurt am Main

Telefonnr.: 069-305-4302

Telefaxnr.: 069-305-16350

Fernschreibnr.:

☒ Zustellanschrift: Dieses Kästchen ist anzukreuzen, wenn kein Anwalt oder gemeinsamer Vertreter bestellt ist und statt dessen im obigen Feld eine spezielle Zustellanschrift angegeben ist.

Feld Nr. V BESTIMMUNG DER STAATEN

Die folgenden Bestimmungen nach Regel 4.9 Absatz a werden hiermit vorgenommen (bitte die entsprechenden Kästchen ankreuzen; wenigstens ein Kästchen muß angekreuzt werden):

Regionales Patent

- ☐ AP ARIPO-Patent: GH Ghana, GM Gambia, KE Kenia, LS Lesotho, MW Malawi, SD Sudan, SZ Swasiland, UG Uganda, ZW Simbabwe und jeder weitere Staat, der Vertragsstaat des Harare-Protokolls und des PCT ist
- ☐ EA Eurasisches Patent: AM Armenien, AZ Aserbaidshan, BY Belarus, KG Kirgisistan, KZ Kasachstan, MD Republik Moldau, RU Russische Föderation, TJ Tadschikistan, TM Turkmenistan und jeder weitere Staat, der Vertragsstaat des Eurasischen Patentübereinkommens und des PCT ist
- ☒ EP Europäisches Patent: AT Österreich, BE Belgien, CH und LI Schweiz und Liechtenstein, CY Zypern, DE Deutschland, DK Dänemark, ES Spanien, FI Finnland, FR Frankreich, GB Vereinigtes Königreich, GR Griechenland, IE Irland, IT Italien, LU Luxemburg, MC Monaco, NL Niederlande, PT Portugal, SE Schweden und jeder weitere Staat, der Vertragsstaat des Europäischen Patentübereinkommens und des PCT ist
- ☐ OA OAPI-Patent: BF Burkina Faso, BJ Benin, CF Zentralafrikanische Republik, CG Kongo, CI Côte d'Ivoire, CM Kamerun, GA Gabun, GN Guinea, GW Guinea-Bissau, ML Mali, MR Mauretanien, NE Niger, SN Senegal, TD Tschad, TG Togo und jeder weitere Staat, der Vertragsstaat der OAPI und des PCT ist (falls eine andere Schutzrechtsart oder ein sonstiges Verfahren gewünscht wird, bitte auf der gepunkteten Linie angeben)

SL Sierra Leone TZ Vereinigte Republik Tansania

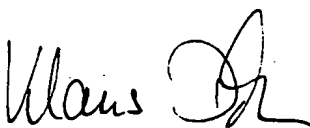
Nationales Patent (falls eine andere Schutzrechtsart oder ein sonstiges Verfahren gewünscht wird, bitte auf der gepunkteten Linie angeben): -

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| <input type="checkbox"/> IN Indien | <input type="checkbox"/> UZ Usbekistan |
| <input type="checkbox"/> IS Island | <input type="checkbox"/> VN Vietnam |
| <input checked="" type="checkbox"/> JP Japan | <input type="checkbox"/> YU Jugoslawien |
| <input type="checkbox"/> KE Kenia | <input type="checkbox"/> ZW Simbabwe |
| <input type="checkbox"/> KG Kirgisistan | |
| <input type="checkbox"/> KP Demokratische Volksrepublik Korea | |
| <input checked="" type="checkbox"/> KR Republik Korea | |
| <input type="checkbox"/> KZ Kasachstan | |
| <input type="checkbox"/> LC Saint Lucia | |
| <input type="checkbox"/> LK Sri Lanka | |
| <input type="checkbox"/> LR Liberia | |

Kästchen für die Bestimmung von Staaten (für die Zwecke eines nationalen Patents), die dem PCT nach der Veröffentlichung dieses Formblatts beigetreten sind:

- ☐ AE Vereinigte arabische Emirate
- ☐ ZA Südafrika
- ☐ TZ Vereinigte Republik Tansania

Erklärung bzgl. vorsorglicher Bestimmungen: Zusätzlich zu den oben genannten Bestimmungen nimmt der Anmelder nach Regel 4.9 Absatz b auch alle anderen nach dem PCT zulässigen Bestimmungen vor mit Ausnahme der im Zusatzfeld genannten Bestimmungen, die von dieser Erklärung ausgenommen sind. Der Anmelder erklärt, daß diese zusätzlichen Bestimmungen unter dem Vorbehalt einer Bestätigung stehen und jede zusätzliche Bestimmung, die vor Ablauf von 15 Monaten ab dem Prioritätsdatum nicht bestätigt wurde, nach Ablauf dieser Frist als vom Anmelder zurückgenommen gilt. (Die Bestätigung einer Bestimmung erfolgt durch die Einreichung einer Mitteilung, in der diese Bestimmung angegeben wird, und die Zahlung der Bestimmungs- und der Bestätigungsgebühr. Die Bestätigung muß beim Anmeldeamt innerhalb der Frist von 15 Monaten eingehen.)

Feld Nr. VI PRIORITÄTSANSPRUCH		<input type="checkbox"/> Weitere Prioritätsansprüche sind im Zusatzfeld angegeben.		
Anmeldedatum der früheren Anmeldung (Tag/Monat)	Aktenzeichen der früheren Anmeldung	Ist die frühere Anmeldung eine:		
		national Anmeldung: Staat	regionale Anmeldung:* regionales Amt	internationale Anmeldung: Anmeldeamt
Zeile (1) 25. Dezember 1998 (25.12.98)	371554/98	JP		
Zeile (2)				
Zeile (3)				
<input type="checkbox"/> Das Anmeldeamt wird ersucht, eine beglaubigte Abschrift der oben in der (den) Zeile(n) _____ bezeichneten früheren Anmeldung(en) zu erstellen und dem internationalen Büro zu übermitteln (nur falls die frühere Anmeldung(en) bei dem Amt eingereicht worden ist(sind), das für die Zwecke dieser internationalen Anmeldung Anmeldeamt ist)				
<p>* Falls es sich bei der früheren Anmeldung um eine ARIPO-Anmeldung handelt, so muß in dem Zusatzfeld mindestens ein Staat angegeben werden, der Mitgliedstaat der Pariser Verbandsübereinkunft zum Schutz des gewerblichen Eigentums ist und für den die frühere Anmeldung eingereicht wurde.</p>				
Feld Nr. VII INTERNATIONALE RECHERCHENBEHÖRDE				
Wahl der internationalen Recherchenbehörde (ISA) (falls zwei oder mehr als zwei internationale Recherchenbehörden für die Ausführung der internationalen Recherche zuständig sind, geben Sie die von Ihnen gewählte Behörde an; der Zweibuchstaben-Code kann benutzt werden)		Antrag auf Nutzung der Ergebnisse einer früheren Recherche; Bezugnahme auf diese frühere Recherche (falls eine frühere Recherche bei der internationalen Recherchenbehörde beantragt oder von ihr durchgeführt worden ist):		
ISA /		Datum (Tag/Monat/Jahr)	Aktenzeichen	Staat (oder regionales Amt)
Feld Nr. VIII KONTROLLISTE; EINREICHUNGSSPRACHE				
Diese internationale Anmeldung enthält die folgende Anzahl von Blättern:		Dieser internationalen Anmeldung liegen die nachstehend angekreuzten Unterlagen bei:		
Antrag	: 3	1. <input checked="" type="checkbox"/> Blatt für die Gebührenberechnung		
Beschreibung (ohne Sequenzprotokollteil)	: 13	2. <input type="checkbox"/> Gesonderte unterzeichnete Vollmacht		
Ansprüche	: 1	3. <input checked="" type="checkbox"/> Kopie der allgemeinen Vollmacht; Aktenzeichen (falls vorhanden):		
Zusammenfassung	: 1	4. <input type="checkbox"/> Begründung für das Fehlen einer Unterschrift		
Zeichnungen	: 3	5. <input checked="" type="checkbox"/> Prioritätsbeleg(e), in Feld Nr. VI durch folgende Zeilennummer gekennzeichnet:		
Sequenzprotokollteil der Beschreibung	:	6. <input type="checkbox"/> Übersetzung der internationalen Anmeldung in die folgende Sprache:		
Blattzahl insgesamt	: 21	7. <input type="checkbox"/> Gesonderte Angaben zu hinterlegten Mikroorganismen oder anderem biologischen Material		
Abbildung der Zeichnungen, die mit der Zusammenfassung veröffentlicht werden soll (Nr.): 1		8. <input type="checkbox"/> Sequenzprotokolle für Nucleotide und/oder Aminosäuren in computerlesbarer Form		
		9. <input type="checkbox"/> Sonstige (einzeln auflisten):		
		Sprache, in der die internationale Anmeldung eingereicht wird: englisch		
Feld Nr. IX UNTERSCHRIFT DES ANMELDERS ODER DES ANWALTS				
Der Name jeder unterzeichnenden Person ist neben der Unterschrift zu wiederholen, und es ist anzugeben, sofern sich dies nicht eindeutig aus dem Antrag ergibt, in welcher Eigenschaft die Person unterzeichnet.				
 Dr. Klaus Doerr (AV-Nr. 37986)				

1. Datum des tatsächlichen Eingangs dieser internationalen Anmeldung:		Vom Anmeldeamt auszufüllen (11. 12. 99)		11 DEC 1999	
3. Geändertes Eingangsdatum aufgrund nachträglich, jedoch fristgerecht eingegangener Unterlagen oder Zeichnungen zur Vervollständigung dieser internationalen Anmeldung:				2. Zeichnungen <input checked="" type="checkbox"/> eingegangen: <input type="checkbox"/> nicht eingegangen:	
4. Datum des fristgerechten Eingangs der angeforderten Richtigstellungen nach Artikel 11(2) PCT:					
5. Internationale Recherchenbehörde (falls zwei oder mehr zuständig sind): ISA /		6. <input type="checkbox"/> Übermittlung des Recherchenexemplars bis zur Zahlung der Recherchegebühr aufgeschoben			

Vom Internationalen Büro auszufüllen
Datum des Eingangs des Aktenexemplars beim Internationalen Büro:

Zusatzfeld Wird dieses Zusatzfeld nicht benutzt, so sollte dieses Blatt dem Antrag nicht beigelegt werden.

1. Wenn der Platz in einem Feld nicht für alle Angaben ausreicht: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. ..." [Nummer des Feldes angeben] und machen die Angaben entsprechend der in dem Feld, in dem der Platz nicht ausreicht, vorgeschriebenen Art und Weise, insbesondere:

- (i) Wenn mehr als zwei Anmelder und/oder Erfinder vorhanden sind und kein "Fortsetzungsblatt" zur Verfügung steht: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. III" und machen für jede weitere Person die in Feld Nr. III vorgeschriebenen Angaben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.
 - (ii) Wenn in Feld Nr. II oder III die Angabe "die im Zusatzfeld angegebenen Staaten" angekreuzt ist: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. II", "Fortsetzung von Feld Nr. III" bzw. "Fortsetzung von Feld Nr. II und Nr. III" und geben den Namen des Anmelders oder die Namen der Anmelder an und neben jedem Namen den Staat oder die Staaten (und/oder ggf. ARIPO-, eurasisches, europäisches oder OAPI-Patent), für die die bezeichnete Person Anmelder ist.
 - (iii) Wenn der in Feld Nr. II oder III genannte Erfinder oder Erfinder/Anmelder nicht für alle Bestimmungsstaaten oder für die Vereinigten Staaten von Amerika als Erfinder benannt ist: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. II", "Fortsetzung von Feld Nr. III" bzw. "Fortsetzung von Feld Nr. II und Nr. III" und geben den Namen des Erfinders oder die Namen der Erfinder an und neben jedem Namen den Staat oder die Staaten (und/oder ggf. ARIPO-, eurasisches, europäisches oder OAPI-Patent), für die die bezeichnete Person Erfinder ist.
 - (iv) Wenn zusätzlich zu dem Anwalt oder den Anwälten, die in Feld Nr. IV angegeben sind, weitere Anwälte bestellt sind: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. IV" und machen für jeden weiteren Anwalt die entsprechenden, in Feld Nr. IV vorgeschriebenen Angaben.
 - (v) Wenn in Feld Nr. V bei einem Staat (oder bei OAPI) die Angabe "Zusatzpatent" oder "Zusatzzertifikat," oder wenn in Feld Nr. V bei den Vereinigten Staaten von Amerika die Angabe "Fortsetzung" oder "Teilfortsetzung" hinzugefügt wird: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. V" und geben den Namen des betreffenden Staats (oder OAPI) an und nach dem Namen jedes solchen Staats (oder OAPI) das Aktenzeichen des Hauptschutzrechts oder der Hauptschutzrechtsanmeldung und das Datum der Erteilung des Hauptschutzrechts oder der Einreichung der Hauptschutzrechtsanmeldung.
 - (vi) Wenn in Feld Nr. VI die Priorität von mehr als drei früheren Anmeldungen beansprucht wird: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. VI" und machen für jede weitere frühere Anmeldung die entsprechenden, in Feld Nr. VI vorgeschriebenen Angaben.
 - (vii) Wenn in Feld Nr. VI die frühere Anmeldung eine ARIPO Anmeldung ist: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. VI" und geben, unter Angabe der Nummer der Zeile, in der die die frühere Anmeldung betreffenden Angaben gemacht sind, mindestens einen Staat an, der Mitglied der Pariser Verbandsübereinkunft zum Schutz des gewerblichen Eigentums ist und für den die frühere Anmeldung erfolgte.
2. Wenn, im Hinblick auf die Erklärung bzgl. vorsorglicher Bestimmungen in Feld Nr. V, der Anmelder Staaten von dieser Erklärung ausnehmen möchte: In diesem Fall schreiben Sie "Bestimmung(en), die von der Erklärung bzgl. vorsorglicher Bestimmungen ausgenommen ist(sind)" und geben den Namen oder den Zweibuchstaben-Code jedes so ausgeschlossenen Staates an.
3. Wenn der Anmelder für irgendein Bestimmungsamt die Vorteile nationaler Vorschriften betreffend **unschädliche Offenbarung oder Ausnahmen von der Neuheitsschädlichkeit** in Anspruch nimmt: In diesem Fall schreiben Sie "Erklärung betreffend unschädliche Offenbarung oder Ausnahmen von der Neuheitsschädlichkeit" und geben im folgenden die entsprechende Erklärung ab.

Feld Nr. IX. Unterschrift des Anmelder oder des Anwalts


Tetsu Yamamoto

PCT COOPERATION TRE

PCT

NOTIFICATION OF THE RECORDING
OF A CHANGE(PCT Rule 92bis.1 and
Administrative Instructions, Section 422)

From the INTERNATIONAL BUREAU

CELANESE VENTURES GMBH
Intellectual Property Group
Industriepark Höchst
Gebäude K 801
65926 Frankfurt
ALLEMAGNE

Date of mailing (day/month/year) 12 April 2001 (12.04.01)	IMPORTANT NOTIFICATION
Applicant's or agent's file reference 1998/F151 PCT	
International application No. PCT/EP99/09831	International filing date (day/month/year) 11 December 1999 (11.12.99)

1. The following indications appeared on record concerning:		
<input checked="" type="checkbox"/> the applicant	<input type="checkbox"/> the inventor	<input type="checkbox"/> the agent <input type="checkbox"/> the common representative
Name and Address AXIVA GMBH D-65926 Frankfurt am Main Germany	State of Nationality DE	State of Residence DE
	Telephone No. 069-305-4302	
	Facsimile No. 069-305-16350	
	Teleprinter No.	
2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:		
<input type="checkbox"/> the person	<input checked="" type="checkbox"/> the name	<input checked="" type="checkbox"/> the address <input type="checkbox"/> the nationality <input type="checkbox"/> the residence
Name and Address CELANESE VENTURES GMBH 65926 Frankfurt Germany	State of Nationality DE	State of Residence DE
	Telephone No. 069-305-4302	
	Facsimile No. 069-305-26600	
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3. Further observations, if necessary: Please also note the change of address for correspondence.		
4. A copy of this notification has been sent to:		
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<input type="checkbox"/> the International Searching Authority	<input checked="" type="checkbox"/> the elected Offices concerned	
<input checked="" type="checkbox"/> the International Preliminary Examining Authority	<input type="checkbox"/> other:	

The International Bureau of WIPO 34, chemin des Colmbettes 1211 Geneva 20, Switzerland	Authorized officer C. Cupello
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

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To:

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United States Patent and Trademark
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Box PCT
Washington, D.C.20231
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in its capacity as elected Office

Date of mailing (day/month/year) 21 August 2000 (21.08.00)	
International application No. PCT/EP99/09831	Applicant's or agent's file reference 1998/F151 PCT
International filing date (day/month/year) 11 December 1999 (11.12.99)	Priority date (day/month/year) 25 December 1998 (25.12.98)
Applicant YAMAMOTO, Tetsu	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
06 July 2000 (06.07.00)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer Nestor Santesso
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION OF THE RECORDING
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Administrative Instructions, Section 422)

To:

CELANESE VENTURES GMBH
Intellectual Property Group
Industriepark Höchst
Gebäude K 801
65926 Frankfurt
ALLEMAGNE

Date of mailing (day/month/year) 12 April 2001 (12.04.01)	IMPORTANT NOTIFICATION
Applicant's or agent's file reference 1998/F151 PCT	
International application No. PCT/EP99/09831	International filing date (day/month/year) 11 December 1999 (11.12.99)

1. The following indications appeared on record concerning:

☒ the applicant ☐ the inventor ☐ the agent ☐ the common representative

Name and Address AXIVA GMBH D-65926 Frankfurt am Main Germany	State of Nationality DE	State of Residence DE
	Telephone No. 069-305-4302	
	Facsimile No. 069-305-16350	
	Teleprinter No.	

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☐ the person ☒ the name ☒ the address ☐ the nationality ☐ the residence

Name and Address CELANESE VENTURES GMBH 65926 Frankfurt Germany	State of Nationality DE	State of Residence DE
	Telephone No. 069-305-4302	
	Facsimile No. 069-305-26600	
	Teleprinter No.	

3. Further observations, if necessary:

Please also note the change of address for correspondence.

4. A copy of this notification has been sent to:

☒ the receiving Office ☐ the designated Offices concerned
☐ the International Searching Authority ☒ the elected Offices concerned
☒ the International Preliminary Examining Authority ☐ other:

<p>The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland</p> <p>Facsimile No.: (41-22) 740.14.35</p>	<p>Authorized officer C. Cupello</p> <p>Telephone No.: (41-22) 338.83.38</p>
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Method for production of polyelectrolyte membranes and fuel cell

Background of the invention

1. Field of the Invention

This invention relates to a method for the production of a polyelectrolyte membrane and to a fuel cell.

2. Related Art

A fuel cell has an electrolyte and a pair of electrodes separated by the electrolyte. In a fuel cell, a fuel such as hydrogen is supplied to one electrode, and an oxidizing agent such as oxygen is supplied to the other electrode. This will convert the chemical energy involving oxidation of the fuel to electric energy. Hydrogen ion (i.e., proton) permeates through the electrolyte while the reaction gases (i.e., hydrogen and oxygen) does not permeate through the electrolyte. Typically, a fuel-cell stack has a plurality of fuel cells, and each of the cells has an electrolyte and a pair of electrodes separated by the electrolyte.

As electrolytes for fuel cells, solids such as polyelectrolyte membranes or liquids such as phosphoric acid are used. Among these, the polyelectrolyte membranes have received attention as the electrolytes for fuel cells in recent years. For example, perfluorosulfonic acid polymers and complexes between basic polymers and strong acids are used as materials for the polyelectrolyte membranes.

The perfluorosulfonic acid polymer, typically, has a structure in which the side chain having a sulfonic acid group (e.g., a side chain having a sulfonic acid group bound to a perfluoroalkylene group) is bound to a perfluorocarbon skeleton (e.g., a copolymer of tetrafluoroethylene and trifluorovinyl). Since the sulfonic acid group can turn into an anion through the dissociation of its hydrogen ion, it shows proton conductivity.

The polyelectrolyte membranes comprising complexes of basic polymers and strong acids have been developed. In International Publication WO96/13872 and its equivalent U.S. Pat. No. 5,525,436, there is disclosed a method for producing a proton conductive polyelectrolyte membrane by immersing a basic polymer such as a polybenzimidazole in a strong acid such as phosphoric acid or sulfuric acid. The fuel cell employing such a polyelectrolyte membrane has the advantage that it can be operated at 100 °C or above.

In *J. Electrochem. Soc.*, Vol. 142, No. 7, 1995, ppL121-L123, it is described that when a polybenzimidazole is immersed in 11M phosphoric acid for at least 16 h,

the polybenzimidazole will be impregnated with five molecules of phosphoric acid per unit.

Further, in International Publication WO97/37396 and its equivalent U.S. Pat. No. 5,716,727, there is described a method for producing a polyelectrolyte membrane by obtaining a solution of polybenzimidazole dissolved in trifluoroacetic acid, next by adding phosphoric acid to the solution, and subsequently by removing the solvent.

All the disclosures of WO96/13872, *J. Electrochem. Soc.*, Vol. 142, No. 7, 1995, ppL121-L123, and WO97/37396 are incorporated into the present specification by reference.

Where the complexes between basic polymers and strong acids are to be put into practical use as the polyelectrolyte membranes for fuel cells, further improvements on their proton conduction are needed.

In addition, where such polyelectrolyte membranes are manufactured, it is required from the standpoint of their production process that the times of immersion of the basic polymers in the strong acids be brief. In *J. Electrochem. Soc.*, Vol. 142, No. 7, 1995, ppL121-L123, a polybenzimidazole is immersed in phosphoric acid for at least 16 h. This is too time-consuming and the production process will prove to be inefficient.

SUMMARY OF THE INVENTION

This invention provides a method for producing a polyelectrolyte membrane, comprising the step of:

immersing a basic polymer in a strong acid having a concentration sufficient to impregnate the basic polymer with six or more strong acid molecules per polymer repeating unit of the basic polymer at a temperature of not less than 35 °C for a period of 5 h or less.

In the invention, the time of immersion is preferably 1 h or less.

Preferably, the strong acid is phosphoric acid; or alternatively, the strong acid is preferably sulfuric acid. It is further preferred that the strong acid be phosphoric acid having a concentration of not less than 80% by weight.

Preferably, the basic polymer is selected from the group consisting of polybenzimidazoles, polypyridines, polypyrimidines, polyimidazoles, polybenzthiazoles, polybenzoxazoles, polyoxadiazoles, polyquinolines, polyquinoxalines, polythiadiazoles, polytetrazapyrenes, polyoxazoles, polythiazoles, polyvinylpyridines, polyvinylimidazoles, and polybenzimidazoles.

This invention provides a fuel cell comprising a plurality of cells, wherein each of the cells is provided with a polyelectrolyte membrane produced by the method described above and a pair of electrodes sandwiching the polyelectrolyte membrane.

In this invention, the immersion time can be shortened to 5 h or less by setting the temperature to 35 °C or above at the time when the basic polymer is immersed in the strong acid. Accordingly, the production process can be made more efficient.

A large quantity of the strong acid can be allowed to impregnate the basic polymer, specifically at the ratio of six or more strong acid molecules per polymer repeating unit of the basic polymer, by adjusting the concentration of the strong acid. Accordingly, the proton conduction of the polyelectrolyte membranes can be improved and the output of fuel cells can be enhanced.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross-sectional illustration of the fuel cell.

FIG. 2 is an enlarged cross section of "A" in FIG. 1.

FIG. 3 is a plot illustrative of the correlation between the concentrations of phosphoric acid and the numbers of phosphoric acid molecules per polymer repeating unit of a polybenzimidazole.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

This invention includes the step of immersing a basic polymer in a strong acid of a predetermined concentration at a temperature of not less than 30 °C for a period of 5 h or less. By carrying out the immersion step at 30 °C or above, it becomes possible to shorten the time needed to immerse the basic polymer in the strong acid: specifically, the time has turned out to be 5 h or less.

The immersion step is to be carried out preferably at 35 °C or above, more preferably at 40 °C or above, in particular preferably at 50°C or above. As the temperature of the immersion step increases, the immersion times can be further shortened.

Thus, by raising the immersion temperature, the immersion time can be made 1 h or less, and can even be made 30 min or less. Shortening the immersion times improves the efficiency of the production process.

However, because the stability of the basic polymers and the safety precautions required to handle the strong acids at high temperatures should be taken into consideration, the immersion step is to be carried out at 200 °C or below, more preferably at 100 °C or below, and most preferably at 80 °C or below.

This invention includes the step of immersing the basic polymer in the strong acid having a concentration sufficient to impregnate the basic polymer with six or more strong acid molecules per polymer repeating unit of the basic polymer. As the concentration of the strong acid increases, the basic polymer can be impregnated with more strong acid. Thus, the impregnation quantity of the strong acid increases; and it improves the proton conduction of a complex between the basic polymer and the strong acid. When the basic polymers are used as the electrolyte membranes for fuel cells, the output of the fuel cells will be enhanced.

It is preferred that the strong acid be in a concentration sufficient to impregnate the basic polymer with eight or more strong acid molecules per polymer repeating unit of the basic polymer. Further, it is more preferred that the concentration be enough to impregnate the basic polymer with nine or more strong acid molecules per polymer repeating unit of the basic polymer.

In WO96/13872 and WO97/37396, a dopant level of not less than 200 molar per cent and that of not less than 300 molar per cent are disclosed, respectively. The former level corresponds to two or more strong acid molecules being present per polymer repeating unit of a basic polymer, and the latter level to three or more strong acid molecules, respectively.

Protic strong acids are used as the strong acid; for example, phosphoric acid and sulfuric acid are preferably used.

As used in the present specification, the "phosphoric acid" includes phosphorous acid (H_3PO_3), orthophosphoric acid (H_3PO_4), pyrophosphoric acid ($\text{H}_4\text{P}_2\text{O}_7$), triphosphoric acid ($\text{H}_5\text{P}_3\text{O}_{10}$), and metaphosphoric acid. The phosphoric acid, particularly orthophosphoric acid, has a concentration of not less than 80% by weight preferably; more preferably, a concentration of not less than 85% by weight; even more preferably, a concentration of not less than 90% by weight; and most preferably, a concentration of not less than 95% by weight. This is because the basic polymer can be impregnated with a larger number of strong acid molecules as the concentration of the strong acid increases.

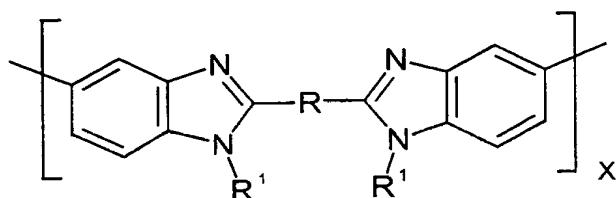
In this invention, the strong acid may be heated to a predetermined temperature, and then, the basic polymer may be immersed in the heated strong acid. Preferably, the basic polymer that has been shaped into a membrane form is immersed in the strong acid. For example, the basic polymer may be shaped into the membrane form following to the doctor blade method.

Alternatively, the basic polymer may be shaped into the membrane form according to the method as described in Japanese Patent Appln. Hei 10-125560, filed May 8, 1998: entitled "Method for Producing Polyelectrolyte Membranes and

Fuel Cells." Specifically, a liquid medium containing not less than 1% by weight of a basic polymer and a solvent having a boiling point or an azeotropic point of from 60°C to 220°C is poured into a cylinder the inner circumference of which has a cylindrical configuration; next, the cylinder is rotated. At that point, the solvent is allowed to evaporate through centrifugation by the rotation; concurrently, a polyelectrolyte membrane having a cylindrical form of almost uniform thickness is formed on the inner circumference of the cylinder. Thereafter, the polyelectrolyte membrane having a cylindrical form is cut out to yield a polyelectrolyte membrane having a film form. This method permits the basic polymer to shape into a uniform matrix within its polyelectrolyte membrane. The disclosure of Japanese Patent Appln. Hei 10-125560 is incorporated into the present specification by reference.

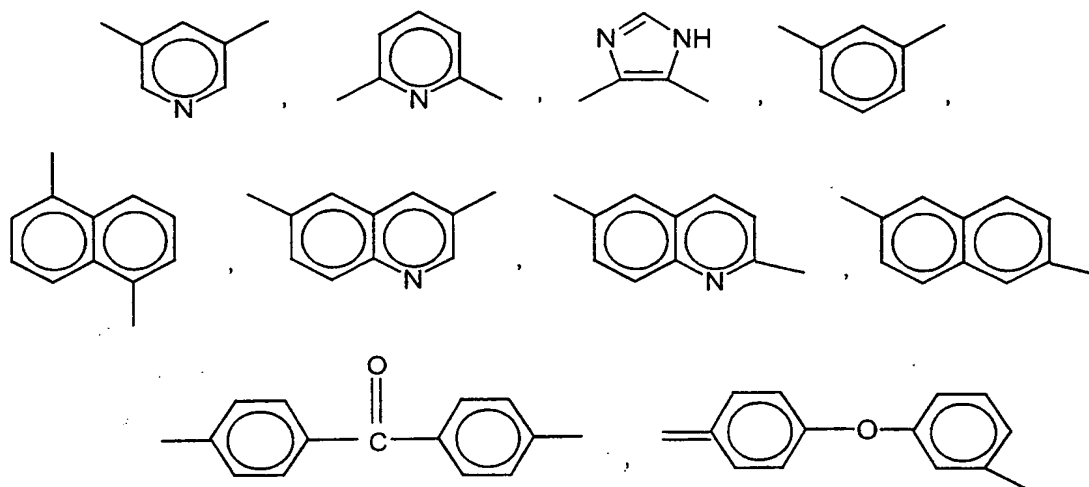
In this invention, basic polymers are used. Such basic polymers include polybenzimidazoles, polypyridines, polypyrimidines, polyimidazoles, polybenzthiazoles, polybenzoxazoles, polyoxadiazoles, polyquinolines, polyquinoxalines, polythiadiazoles, polytetrazapyrenes, polyoxazoles, polythiazoles, polyvinylpyridines, polyvinylimidazoles, polybenzimidazoles, etc. Among these, polybenzimidazoles are preferred. The basic polymers described in WO96/13872 are also preferably used.

As the polybenzimidazoles, preferably usable are, for example, those of the following formula:



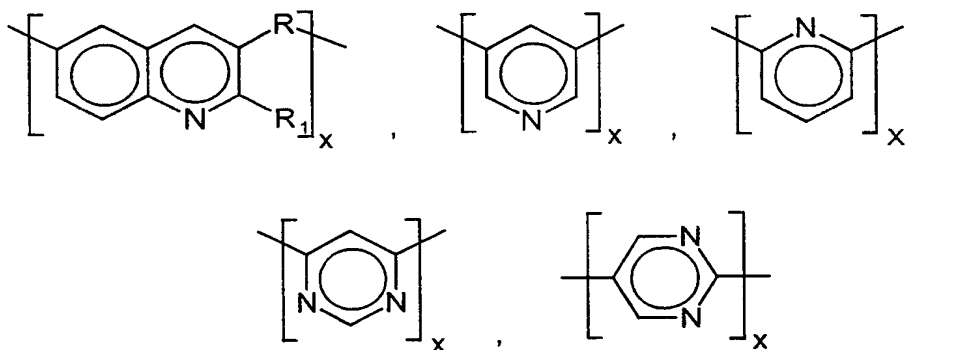
wherein R represents alkylene, perfluoroalkylene, or a substituent of any of the following formulae:

6



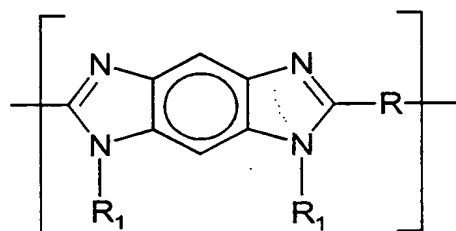
further wherein each of alkylene and perfluoroalkylene groups, which may be R, has from 1 to 10 carbons preferably, and more preferably from 1 to 6 carbons, and still further wherein R¹ may be the same or different and represents hydrogen, alkyl or phenyl, wherein the alkyl preferably has from 1 to 6 carbons and is optionally substituted with halogen, sulfone, or the like.

The basic polymers which may also be used are represented by the following formula:



wherein R and R¹ are as previously defined.

Furthermore, the basic polymers which may also be used are polybenzobisimidazoles of the following formula:



wherein R and R¹ are as previously above.

The polyelectrolytes obtained by this invention, viz. the complexes between the basic polymers and the strong acids, are proton conductive; therefore, they can preferably be used as the electrolytes for cells. Nevertheless, the polyelectrolytes are not be limited to be of use for cells; but they can also be used as the electrolytes for display elements, electrochromic elements or various sensors.

According to another aspect of this invention, the polyelectrolyte membranes can preferably be used in the cells for fuel cells.

In FIG. 1, a cell 10 of a fuel cell is provided with an electrolyte membrane 12 and a pair of electrodes 20 sandwiching the electrolyte membrane 12. The electrode 20 is provided with a catalyst layer 14 conducting electrode reaction and with a gas diffusion layer 22 for supplying the catalyst layer 14 with a reaction gas.

In FIG. 2, the catalyst layer 14 is provided with a matrix 15 comprising an electrolyte membrane and with two or more catalyst particles 16 dispersed in the matrix. The matrix 15, together with the electrolyte membrane 12, forms a hydrogen ion-conducting channel. Preferably, the material for the matrix 15 is identical to the material for the electrolyte membrane 12. However, these materials may be different from each other. The matrix 15 may be porous so that the reaction gas can pass through. The catalyst particles 16 are preferably in contact with each other: this forms an electron-conducting channel.

Each of the catalyst particles 16 is provided with a conductive carrier 17 and a catalyst substance 18 supported on the surface of the conductive carrier 17. For example, particles comprising carbon are used as the conductive carrier 17. Simple substance of platinum, alloys of platinum, and the like are used as the catalyst substance 18. In FIG. 2 the catalyst substance 18 coats the surface of the conductive carrier 17, but it may be in a particulate form.

The gas diffusion layer 22 is porous so that the reaction gas can be allowed to diffuse. In FIG. 2 the gas diffusion layer 22 comprises two or more conductive particles 26 that form a gap 24. For example, particles comprising carbon are used as the conductive particles 26, and may be the same as the conductive carrier 17.

Conductive substances such as carbon fiber may be used in place of the conductive particles 26.

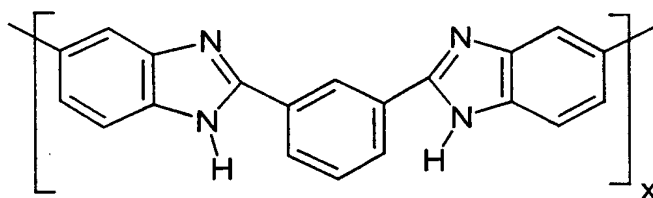
The polyelectrolytes of this invention can be used as the electrolyte membrane 12. Thus a cell precursor having the electrolyte membrane 12 and either or both of the catalyst layers 14 can also be prepared. Moreover, a cell can then be produced by fixing the gas diffusion layer 22 to such a precursor.

EXAMPLES

The following examples are merely illustrative of this invention, and are not to be construed as limitations thereof.

Reference Example

Polybenzimidazole having the structural formula described below and having an intrinsic viscosity of 1.1 (available from Hoechst Celanese Inc.) was dissolved in N,N-dimethylacetamide to yield a solution having a resin concentration of 5.0% by weight.



This solution, 83g, was poured into a tubular cylinder made of stainless steel (141 mm in inner diameter and 408 mm long), and it was rotated at 1100 rpm and at 90 °C for 2 h to yield a polybenzimidazole membrane in a cylindrical form. When the thickness of the resulting polybenzimidazole membrane was measured at arbitrary 6 points, its mean membrane thickness was 30.2 μm ; the deviations of the maximum value of measurement and the minimum value of measurement from the mean membrane thickness are within 1 μm .

EXAMPLE 1

This polybenzimidazole was impregnated with orthophosphoric acid. The polybenzimidazole membrane (30 μm thick) was cut out in 3-cm square pieces. The films were washed with water to wash away the remaining N,N-dimethylacetamide. Then, they were dried at reduced pressure and the weights of the films were measured.

Thereafter, the dried polybenzimidazole films were placed in sample vials. To these was added each 30 ml of 85% by weight aqueous orthophosphoric acid, and immersion was carried out at temperatures and for periods of time as listed in Table

1. After lapses of the predetermined times, the polybenzimidazole films impregnated with orthophosphoric acid were removed from the phosphoric acid and excess phosphoric acid on their surfaces was thoroughly wiped off with filter papers. Subsequently, the weight increments were determined by weighing. After weighing, the polybenzimidazole films were placed in 1-l volumetric flasks, and deionized water was filled up to the measuring lines and stirred. Orthophosphoric acid was extracted from the polybenzimidazole films to obtain aqueous phosphoric acid solutions. The aqueous phosphoric acid solutions thus obtained were titrated with 0.02 N sodium hydroxide solution, and the quantities of orthophosphoric acid having impregnated the polybenzimidazole films were determined. The difference between the weight increment after impregnation with orthophosphoric acid and the weight of orthophosphoric acid of impregnation was calculated to be the quantity of the water that had been adsorbed to each polybenzimidazole film impregnated with orthophosphoric acid. These results are shown in Table 1.

Table 1

Sample	Temperature (°C) of phosphoric acid	immersion time	dried weight of PBI film (g)	film weight after impreg- nation with phosphoric acid (g)	impregnation quantity of phosphoric acid (g)	quantity of adsorbed water (g)	number of phosphoric acid molecules of impregnation per polymer repeating unit	number of adsorbed water molecules per polymer repeating unit
1	50	5 min	0.0692	0.3739	0.2017	0.088	9.58	21.69
2	50	10 min	0.0692	0.3739	0.2217	0.088	10.08	18.52
3	50	30 min	0.0720	0.3744	0.2181	0.084	9.53	19.22
4	40	15 min	0.0812	0.4309	0.2524	0.097	9.78	20.55
5	40	20 min	0.0713	0.3770	0.2181	0.088	9.62	21.07
6	40	30 min	0.0741	0.3772	0.2181	0.085	9.26	19.66
7	40	55 min	0.0698	0.3598	0.2107	0.079	9.49	19.44
8	40	5 h	0.0711	0.3874	0.2279	0.088	10.09	21.33
9	23	1 h	0.0722	0.3285	0.1911	0.065	8.33	15.47
10	23	2 h	0.0745	0.3902	0.2328	0.083	9.83	19.08
11	23	3 h	0.0701	0.3688	0.2156	0.083	9.68	20.32
12	23	5 h	0.0707	0.3689	0.2106	0.088	9.38	21.24
13	23	24 h	0.0581	0.3281	0.1960	0.074	10.61	21.79

From Table 1, it is understood that the use of 85% by weight orthophosphoric acid at 40 °C can remarkably shorten the time during which the impregnation quantity of orthophosphoric acid reaches its equilibrium. Especially, impregnation at 50 °C can shorten the immersion times to approximately one hundredth of the 16 h in a known method.

EXAMPLE 2

According to the method of Example 1, the impregnation of orthophosphoric acid was carried out at 23 °C for 24 h by varying the concentration of orthophosphoric acid within 50-85% by weight. Thus, the relationship between the impregnation quantities of phosphoric acid and the phosphoric acid concentrations was examined. These results are shown in Table 2 and FIG. 3.

Table 2

Phosphoric acid concentration (% by weight)	Dried weight of PBI film (g)	film weight after impregnation with phosphoric acid (g)	impregnation quantity of phosphoric acid (g)	quantity of adsorbed water (g)	number of phosphoric acid molecules of impregnation per polymer repeating unit	number of adsorbed water molecules per polymer repeating unit
85	0.0581	0.3281	0.196	0.07396	10.61	21.79
80	0.0590	0.2457	0.137	0.04952	7.32	14.39
70	0.0609	0.2006	0.108	0.03192	5.57	8.98
60	0.0604	0.1724	0.086	0.0257	4.49	7.30
50	0.0587	0.1511	0.072	0.0206	3.85	6.00

From Table 2, it is understood that the higher the concentration of orthophosphoric acid is, the greater the impregnation quantity of phosphoric acid in the polybenzimidazole becomes. This correlation holds not only at room temperature, but also under warming at 40 ° and 50 °C.

EXAMPLE 3

A polybenzimidazole membrane having a thickness of 50 μm was produced according to the method of Reference Example. This polybenzimidazole membrane was immersed in 85% by weight phosphoric acid at 40 °C for 1 h to yield a polyelectrolyte membrane. This polyelectrolyte membrane was cut out in a circular piece of 7-cm diameter. Next, it was sandwiched by two sheets of carbon electrodes for a fuel cell of the polyelectrolyte type, which were commercially available, and hotpressed to yield a cell for fuel battery. When hydrogen and air were introduced into this cell and electricity was generated, a very high output of was obtained: 350 mW/cm^2 at 160 °C and 0.5 V under 1 atmosphere, and 650 mW/cm^2 at 160 °C and 0.5 V under 3 atmospheres, respectively.

CLAIMS

1. A method for producing a polyelectrolyte membrane, comprising the step of:
5 immersing a basic polymer in a strong acid having a concentration sufficient to impregnate the basic polymer with six or more strong acid molecules per polymer repeating unit of the basic polymer at a temperature of not less than 30 °C for a period of 5 h or less.
- 10 2. The method according to claim 1, wherein the immersion time is 1 hour or less.
3. The method according to claim 1 or claim 2, wherein the strong acid is phosphoric acid.
- 15 4. The method according to claim 1 or claim 2, wherein the strong acid is sulfuric acid.
5. The method according to claim 1 or claim 2, wherein the strong acid is
20 phosphoric acid having a concentration of not less than 80% by weight.
6. The method according to claim 1 or claim 2, wherein the basic polymer is selected from the group consisting of polybenzimidazoles, polypyridines, polypyrimidines, polyimidazoles, polybenzthiazoles, polybenzoxazoles,
25 polyoxadiazoles, polyquinolines, polyquinoxalines, polythiadiazoles, polytetrazapyrenes, polyoxazoles, polythiazoles, polyvinylpyridines, polyvinylimidazoles, and polybenzimidazoles.
7. A fuel cell comprising a plurality of cells, wherein the cell is provided with a
30 polyelectrolyte membrane produced by the method according to any of claims 1-6 and with a pair of electrodes sandwiching the polyelectrolyte membrane.

1 / 3

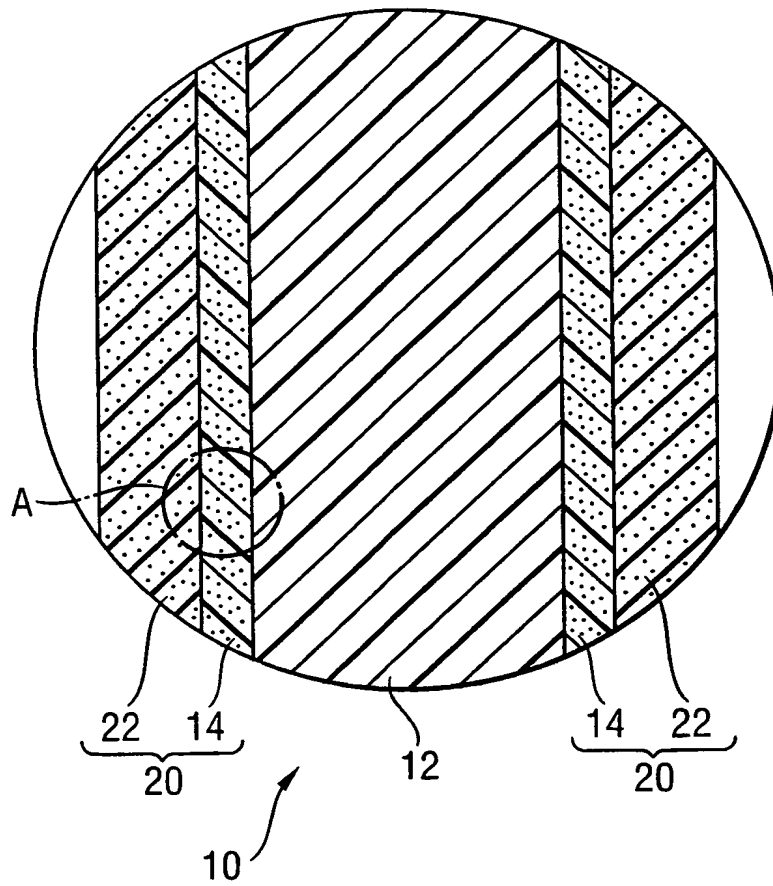


Fig. 1

2 / 3

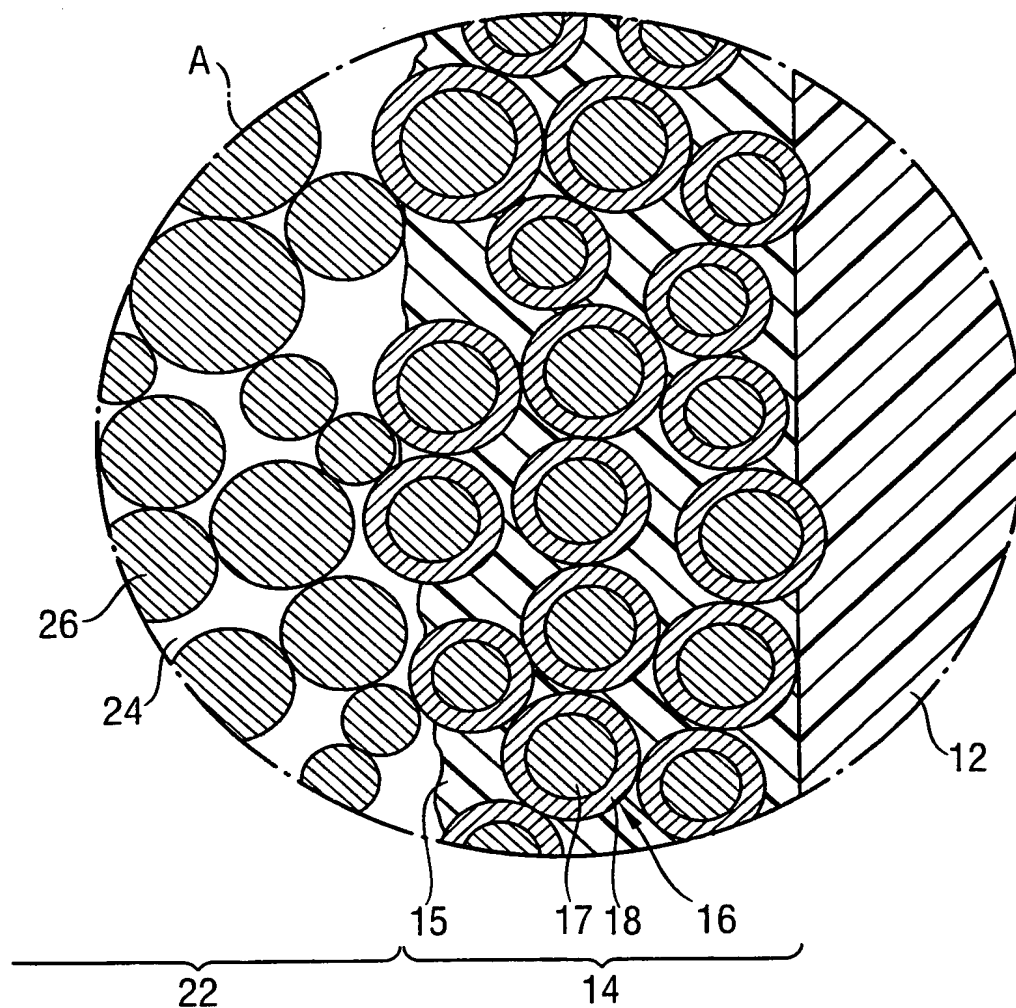
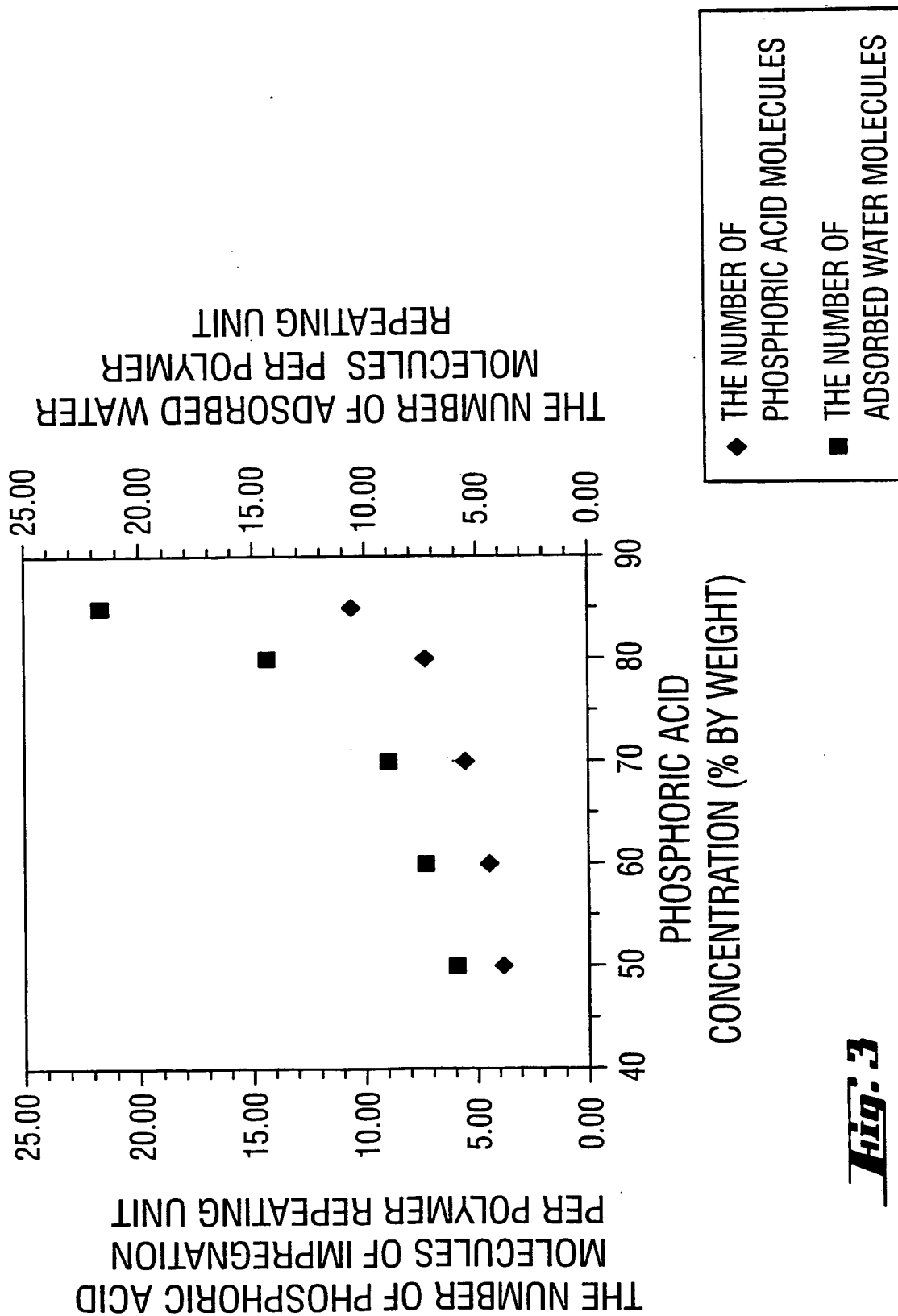


Fig. 2

3 / 3

**Fig. 3**

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/EP 99/09831

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08J5/22 C25B9/00 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J C25B H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 14505 A (HOECHST CELANESE CORP ;SANSONE MICHAEL J (US); MARIKAR FARUQ (US);) 9 April 1998 (1998-04-09) example 1 page 7, line 12 -page 8, line 2 claims 1,10,12	1-7
A	WAINRIGHT J S ET AL: "ACID-DOPED POLYBENZIMIDAZOLES: A NEW POLYMER ELECTROLYTE" JOURNAL OF THE ELECTROCHEMICAL SOCIETY,US,ELECTROCHEMICAL SOCIETY. MANCHESTER, NEW HAMPSHIRE, vol. 142, no. 7, 1 July 1995 (1995-07-01), pages 121-123, XP000770321 ISSN: 0013-4651 cited in the application the whole document	1

-/-



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents:

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- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *Z* document member of the same patent family

Date of the actual completion of the international search

11 May 2000

Date of mailing of the international search report

22/05/2000

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INTERNATIONAL SEARCH REPORT

International Application No.

PCT/EP 99/09831

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>WANG J -T ET AL: "A H₂/O₂ FUEL CELL USING ACID DOPED POLYBENZIMIDAZOLE AS POLYMER ELECTROLYTE"</p> <p>ELECTROCHIMICA ACTA,GB,ELSEVIER SCIENCE PUBLISHERS, BARKING, vol. 41, no. 2, 1 February 1996 (1996-02-01), pages 193-197, XP000770320 ISSN: 0013-4686 page 193, left-hand column, line 1 -page 195, left-hand column, line 8</p>	1
A	<p>US 4 795 536 A (YOUNG PING ET AL) 3 January 1989 (1989-01-03) column 7, line 35 -column 8, line 40 claim 1</p>	1
A	<p>WO 97 37396 A (UNIV CASE WESTERN RESERVE) 9 October 1997 (1997-10-09) cited in the application claims 1-4</p>	1
A	<p>WO 96 13872 A (UNIV CASE WESTERN RESERVE) 9 May 1996 (1996-05-09) cited in the application claims 34-36</p>	1
A	<p>US 5 599 639 A (OGATA NAOYA ET AL) 4 February 1997 (1997-02-04) claims 1,2,9</p>	1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/09831

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